Tritium inventory in a $LiAlO_2$ blanket

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Abstract

Although lithium ceramic materials such as Li_2O, $LiAlO_2$, Li_2ZrO_3 and Li_4SiO_4 are considered as breeding materials in the blanket of a D–T fusion reactor, the release behavior of the bred tritium in these solid breeder materials has not been fully understood. Most of the results of in situ tritium release experiments are analyzed assuming that the overall release process of tritium is mainly controlled with the process of tritium diffusion in the crystal grain of a solid breeder material. However, it has been pointed out by many authors that contribution of the surface reaction cannot be ignored. Effects of diffusion of tritium in the grain, absorption of water in the bulk of grain and adsorption of water on the surface of grain together with isotope exchange reactions on the tritium inventory under the steady state condition are discussed in this study. Comparison of the estimated tritium inventory using the way of this study with the data obtained in several in situ experiments shows that good agreements are obtained when the effective diffusivity obtained from the data by Kwast et al. is used. The better agreements are obtained when existence of some water vapor is assumed in the purge gas. © 1997 Elsevier Science B.V.

1. Introduction

In order to understand the release behavior of tritium bred in a solid breeder material, it is necessary to know the contribution of such mass transfer steps as

(1) diffusion of tritium in crystal grain,

(2) effect of radiation defects on tritium migration in crystal grain,

(3) adsorption of tritium on grain surface,

(4) absorption of tritium in crystal grain,

(5) amount of isotope exchange capacity besides adsorption capacity,

(6) isotope exchange reaction between gaseous hydrogen, H_2, in the gas stream and tritium on grain surface,

(7) isotope exchange reaction between water vapor, H_2O, in the gas stream and tritium on grain surface,

(8) water formation reaction at addition of H_2 to the blanket purge gas,

(9) transfer of hydrogen isotopes and water through pores of the sintered pellet,

(10) transfer of hydrogen isotopes and water through the boundary layer formed on the surface of the sintered pellet to the gas stream,

together with the system effect consisting of

(11) adsorption of tritium on the piping surface,

(12) isotope exchange reaction between gaseous tritium, HT or T_2, in gas stream and water or -OH bases on the piping surface,

(13) isotope exchange reaction between tritiated water in the gas stream and water or -OH bases on the piping surface,

(14) permeation of tritium through piping materials,

(15) absorption of tritium in piping materials,

(16) flow condition of purge gas in a sub-unit constituting the piping system,

and the tritium behaviors in the monitor system such as

(17) memory effect,

(18) Jesse effect.

Adsorption or desorption and two types of isotope exchange reactions contribute as the surface reactions to

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the tritium transfer. Most results of the in situ experiments so far are analyzed by assuming that the overall release process of tritium is mainly controlled with the process of diffusion in the crystal grain. However, it has also been pointed out recently that the contribution of the process of surface reactions cannot be ignored [1–4]. The present authors have quantified the amount of water captured in Li_2O, $LiAlO_2$, Li_2ZrO_3 and Li_4SiO_4 and compared the tritium inventory at the blanket due to sorption with that due to diffusion in the crystal grain in the previous papers [5–8]. This comparison shows that the inventory due to sorption is much larger than that due to diffusion unless otherwise a large amount of H_2 or water vapor is swamped to the purge gas in the high temperature blanket. It has been also pointed out by the present authors that some -OH bases (the strongly bound chemically adsorbed water or crystal water formed on the grain surface of a solid breeder material at the pellet manufacturing process) can act as the tritium trap through isotope exchange reactions [9].

Discussions about tritium inventory in the $LiAlO_2$ blanket under the uniform and steady state condition are performed in this study by considering the processes of diffusion, absorption, adsorption and isotope exchange reactions. Comparison of estimated tritium inventory in this study with data in several in situ experiments such as EXOTIC-6 [10], LILA-3 [11], TEQUILA-1 [12], CORELLI-2 [13], LISA-2 [2] or MOZART [14] experiment are also performed.

2. Inventory estimation method

Contribution of the process of tritium diffusion in the crystal grain, absorption of water into bulk of lithium ceramic, adsorption of water onto the grain surface, isotope exchange reaction between H_2 in the purge gas and tritium on the grain surface or isotope exchange reaction between H_2O in the purge gas and tritium on the grain surface are considered in estimation of the tritium inventory in a uniform blanket under the steady state condition.

The diffusion inventory in the blanket packed with solid breeder particles made of spherical grain is given as

$$I_D = G_T d_p^2 / 60 D_T \ (\text{mol T}), \tag{1}$$

where G_T is the tritium generation rate in the whole blanket (mol/s), d_p the grain diameter (m), and D_T the effective diffusivity of tritium in grain (m²/s).

Then, the average residence time under diffusion of tritium in the crystal grain, I_D / G_T, is given as

$$\theta_D = d_p^2 / 60 D_T \ (\text{s}). \tag{2}$$

If the diffusion in the crystal grain is the rate controlling step, various parameters in the purge gas such as partial pressure of hydrogen or water vapor do not affect the residence time of tritium.

The adsorption inventory is given as the following equation where the effectiveness correction factor α_{ex} shows the relative value of the isotope exchange reaction rate between H_2 and tritium on the grain surface to the isotope exchange reaction rate between H_2O and tritium on the grain surface because it is possible that both reaction rates are different to each other.

$$I_{ad} = Q_{ad} M P_{T_2O} / \{ (P_{H_2} / \alpha_{ex}) + P_{T_2O} + P_{H_2O} $$
$$+ P'_{H_2O} \} \ (\text{mol T}), \tag{3}$$

where P_{H_2} is the partial pressure of H_2 in the purge gas (Pa), P_{H_2O} the partial pressure of H_2O swamped to the purge gas (Pa), P'_{H_2O} the partial pressure of residual H_2O in the purge gas (Pa), M the amount of Li ceramics in blanket (mol), and α_{ex} the effectiveness correction factor (—).

The partial pressure of tritium in the purge gas, P_{T_2O}, is given as

$$P_{T_2O} = P_{He} G_T / G_{He} \ (\text{Pa}), \tag{4}$$

where all tritium bred in the grain is considered to be released in the chemical form of water and P_{He} (Pa) and G_{He} (mol/s) are total pressure of helium purge gas and flow rate, respectively.

The amount of water adsorption, Q_{ad}, is given as

$$Q_{ad} = C_{ad} A_{BET} P^n \exp(-E_{ad} / RT) \ (\text{mol/mol}) \tag{5}$$

in the previous papers [5–8], where A_{BET} ($= 6 / \rho d_p$; ρ: density (g/m³)) is the BET surface area of the breeder particle and the total partial pressure of water in various form is given as

$$P = P_{T_2O} + P_{H_2O} + P'_{H_2O} \ (\text{Pa}), \tag{6}$$

where HTO is treated as a mixture of $\frac{1}{2}T_2O$ and $\frac{1}{2}H_2O$ in this study following the way taken in consideration of tritium behavior in a catalyst bed or a adsorption bed by the present authors [15].

Then, the average residence time due to the process of adsorption is given as follows:

$$\theta_{ad} = C_{ad}(M P_{He} / G_{He}) A_{BET}$$
$$\times \frac{(P_{T_2O} + P_{H_2O} + P'_{H_2O})^n}{(P_{H_2} / \alpha_{ex}) + P_{T_2O} + P_{H_2O} + P'_{H_2O}}$$
$$\exp(-E_{ad} / RT) \ (\text{s}). \tag{7}$$

The absorption inventory is given as

$$I_{ab} = Q_{ab} M P_{T_2O} / (P_{T_2O} + P_{H_2O} + P'_{H_2O}) \ (\text{mol T}), \tag{8}$$

where no absorption of H_2 into bulk of lithium ceramic is considered.

The amount of water absorbed into lithium ceramic is given as follows as reported in the previous paper for Li_2O [5].

$$Q_{ab} = C_{ab} p^m \exp(-E_{ab} / RT) \ (\text{mol/mol}). \tag{9}$$

Then, the average residence time due to the process of absorption is given as

$$\theta_{ab} = C_{ab}(MP_{He}/G_{He})\left(P_{T_2O} + P_{H_2O} + P'_{H_2O}\right)^{m-1}$$
$$\exp(-E_{ab}/RT) \;(\text{s}), \tag{10}$$

The tritium inventory due to the isotope exchange capacity is given as

$$I_{ex} = Q_{ex}MP_{T_2O}/\left\{\left(P_{H_2}/\alpha_{ex}\right) + P_{T_2O} + P_{H_2O}\right.$$
$$\left. + P'_{H_2O}\right\} \;(\text{mol T}), \tag{11}$$

where Q_{ex} (mol/mol) is the isotope exchange capacity on the grain surface, which is different from the adsorbed water, and is probably due to the –OH bases strongly bound to the grain surface. Q_{ex} is also proportional to the BET surface area.

The average residence time due to the isotope exchange capacity is given as

$$\theta_{ex} = Q_{ex}(MP_{He}/G_{He})/\left\{\left(P_{H_2}/\alpha_{ex}\right) + P_{T_2O} + P_{H_2O}\right.$$
$$\left. + P'_{H_2O}\right\} \;(\text{s}). \tag{12}$$

The mass transfer resistance at the transfer of tritium through the pores of sintered pellets and that through the boundary layer are considered to be negligibly small in this study.

Then, the total inventory in the blanket is given as

$$I_{Total} = I_D + I_{ad} + I_{ab} + I_{ex} \;(\text{mol T}), \tag{13}$$

and the total average residence time is given as

$$\theta_{Total} = \theta_D + \theta_{ad} + \theta_{ab} + \theta_{ex} \;(\text{s}). \tag{14}$$

As can be seen from the above equations, the effects of the tritium generation rate, temperature, grain size, partial pressure of H_2, partial pressure of water vapor or flow rate of the purge gas on each tritium inventory are different. Accordingly it is necessary to understand all mass transfer steps dominating the tritium migration behavior before estimation of the tritium inventory in the blanket operated under a certain condition.

It is assumed in estimation of the tritium inventory using the model of this study that the equations for adsorption capacity, isotope exchange capacity and effective diffusivity are applicable in the wide range of temperature or partial pressure in spite of the ranges in experiments.

3. Results and discussions

3.1. Tritium diffusivity in LiAlO₂

In estimation of the diffusion inventory under the irradiation condition, it is necessary to obtain the reliable effective diffusivity of tritium in the crystal grain which includes the effect due to the formation of irradiation defects. The diffusivities by various authors for LiAlO₂,

however, do not agree with each other as shown in Fig. 1 [1,2,10,11,16–18,21–23]. Insufficient correction of the effect due to surface reactions, the system effect or the effect due to irradiation defects in estimation of diffusivity may be the reason of the large discrepancy.

The total average residence time measured by Kwast et al. [10] shown in Fig. 2 is considered to be taken in the range where diffusion of tritium in LiAlO₂ grain is the rate controlling step because the partial pressure of H_2 or H_2O in the purge gas gives no effect on the residence time only in the diffusion controlling step when the tritium transfer model of this study is applied. As can be also seen from this figure, the diffusivity by Kudo and Okuno [16] is too large and that by Kurasawa et al. [1] is too small to correlate the residence time by Kwast et al. [10]. Using the data by Kwast et al. [10], the following equation is presented in this study representing the effective diffusivity of tritium in the crystal grain of LiAlO₂ under the condition of in situ tritium release experiments.

$$D_T = 12.2 \times 10^{-4} \exp(-217\,\text{kJ}/RT)\;(\text{m}^2/\text{s}). \tag{15}$$

This equation gives almost same diffusivity at 873 K as the correlative equation by Botter et al. [17] or that by Tanaka et al. [18], although the temperature dependency is somewhat different.

In estimation of I_{ad}, the values for n, C_{ad} and E_{ad} are taken from our previous work [6] as $\frac{1}{2}$, 2.6×10^{-7} mol g/mol m₂ Pa$^{1/2}$ and 32.2 kJ/mol, respectively.
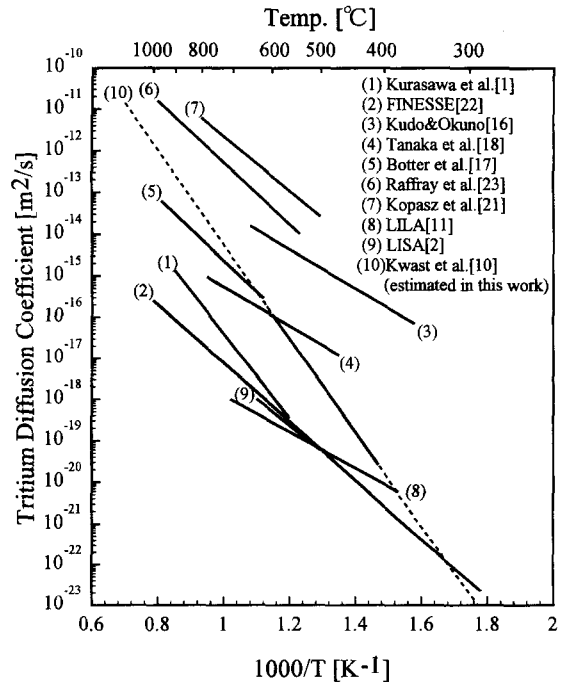

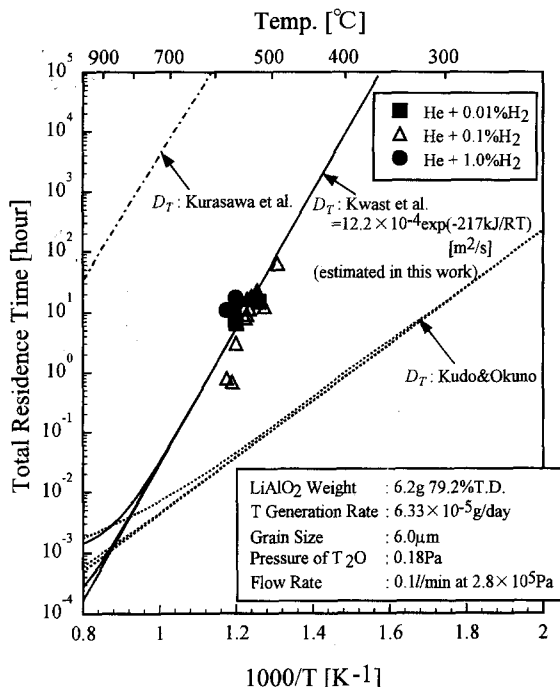Fig. 1. Diffusivity of tritium in the crystal grain of LiAlO₂.

Fig. 2. Comparison of estimated values with observed residence time at in situ experiment (EXOTIC-6).

3.2. Average residence time

In our experiments for $LiAlO_2$, occurrences of absorption of HT or HTO and adsorption of HT have not been observed yet. Accordingly, I_{ab} is excluded in consideration of this study.

The isotope exchange capacities obtained for $LiAlO_2$

previously dried under N_2 gas stream at 673 K for 6 h are shown by the following equation [9]:

$$Q_{ex} = \frac{2.7 \times 10^{-15} A_{BET} \exp(144 \text{ kJ}/RT)}{1 + 2.2 \times 10^{-12} \exp(144 \text{ kJ}/RT)} \text{ (mol/mol)}.$$

(16)

Values of the total average residence time for $LiAlO_2$ blanket purged with 100 kPa He with 100 Pa H_2 estimated using diffusivity by Kwast et al. [10] and are three-dimensionally shown in Fig. 3 where operation of a 1 GWe D–T fusion reactor (overall thermal conversion efficiency: 30%) with the $LiAlO_2$ blanket having 70 tons of Li is supposed. Then, the total residence time of 1 h corresponds to the tritium inventory of 16.7 g in the whole blanket. The contribution of the isotope exchange capacity to the tritium inventory becomes negligibly small when the temperature of the $LiAlO_2$ blanket is higher than 823 K even at so low vapor pressure as 0.1 Pa in the purge gas.

The effect of temperature on the residence time obtained when the diffusivity by Kwast et al. [10] is used is compared with the residence time obtained when the diffusivity by Kudo and Okuno is used in Figs. 4 and 5 also for the blanket purged with He with 100 Pa H_2.

It is known from these figures that effect of the diffusivity cannot be excluded in estimation of the tritium inventory in the $LiAlO_2$ blanket purged by He with 100 Pa H_2 even when the grain size is as small as a few microns if the diffusivity from Kwast et al. [10] is proper, although contribution of diffusion can be ignored even when the grain size is more than 10 microns if the diffusivity by Kudo and Okuno is proper. The effect of adsorption or isotope exchange capacity can be ignored and addition of hydrogen to the purge gas becomes meaningless even when the grain size is smaller than 1 micron if the
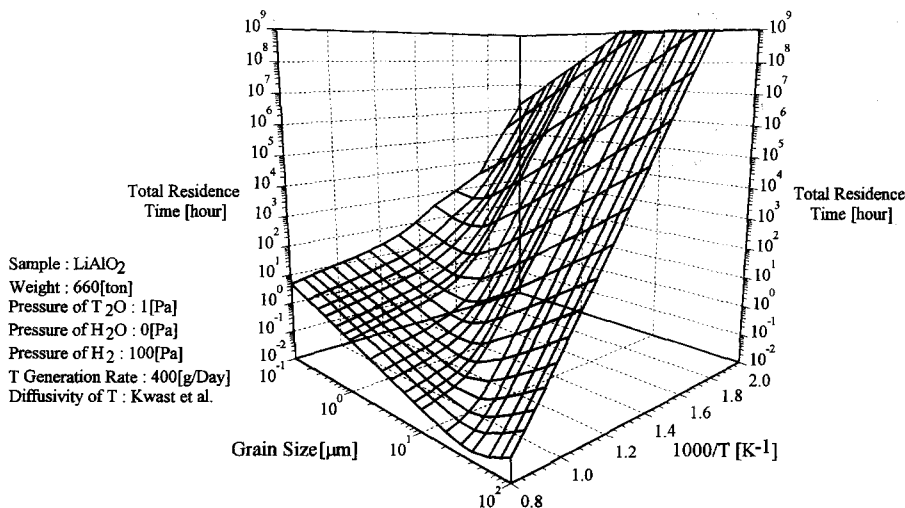

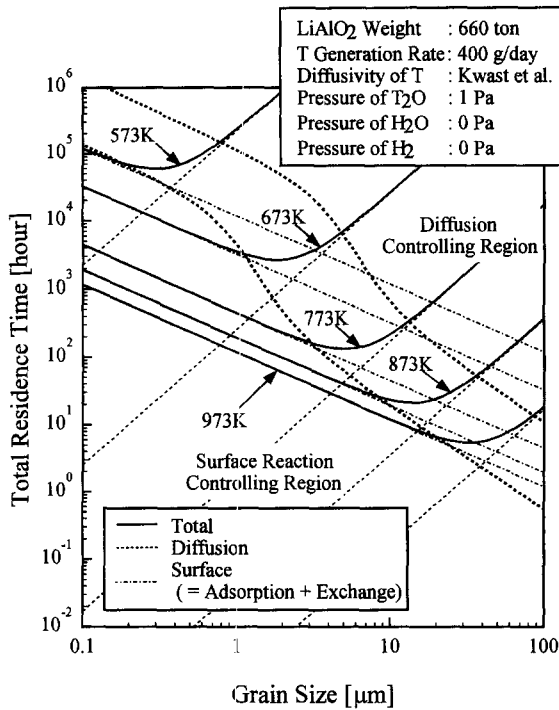Fig. 3. Estimated values of total residence time for $LiAlO_2$ blanket.

Fig. 4. Effect of temperature on residence time for $LiAlO_2$ blanket (He + 100 Pa H_2 purge, D_T: Kwast et al. [10]).
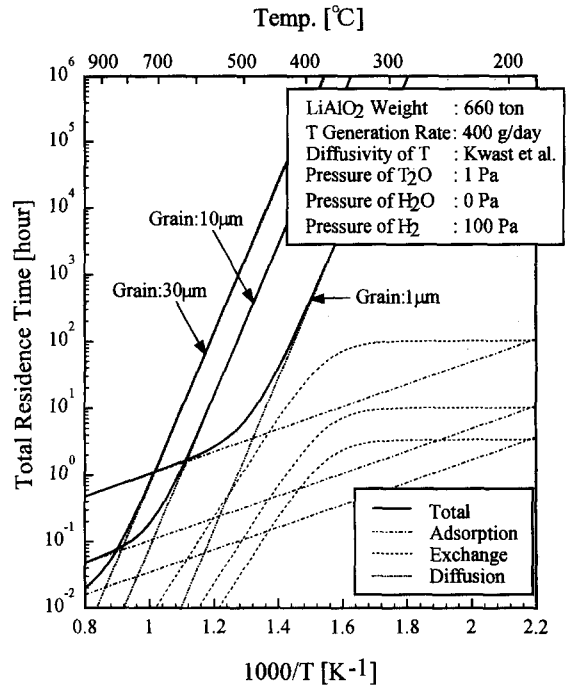

Fig. 6. Effect of grain size on residence time for $LiAlO_2$ blanket (He purge, D_T: Kwast et al. [10]).
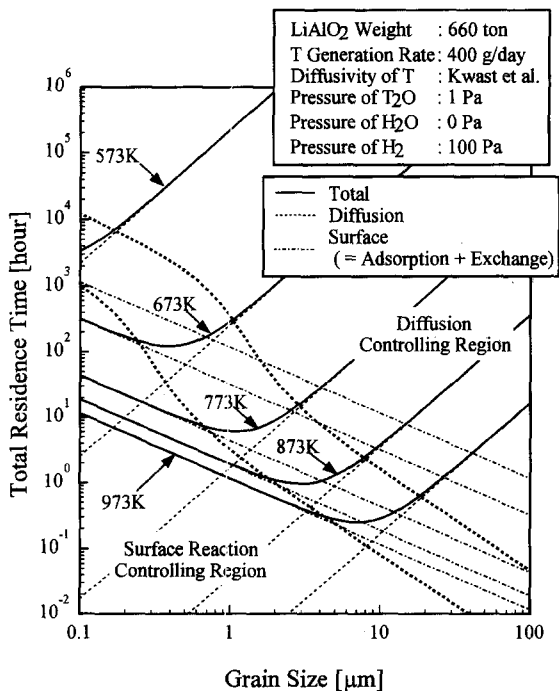

Fig. 5. Effect of temperature on residence time for $LiAlO_2$ blanket (He + 100 Pa H_2 purge, D_T: Kudo and Okuno [16]).

diffusivity by Kurasawa et al. is proper. Accordingly, it is necessary to determine the proper effective diffusivity for estimation of tritium inventory in the blanket under irradiation and consequently for decision of operational conditions of the tritium recovery system. The present authors recommend the diffusivity shown by Eq. (15) as the effective diffusivity of tritium in $LiAlO_2$ under the irradiation condition because the residence time estimated using this equation gives good agreement with the residence time measured in various in situ experiments as compared later.

Effects of the grain size on the residence times when temperature is used as parameter are shown in Figs. 6 and 7 using the diffusivity from Kwast et al. [10].

The region in which more than 90% of the total inventory is occupied by the diffusion inventory is named the diffusion controlling region in this study. Then, there exist a diffusion controlling region and a surface reaction controlling region with a rather wide intermediate region as shown in these figures. Because the effects of tritium generation rate, temperature, grain size, partial pressure of H_2, partial pressure of water vapor or flow rate of purge gas on the tritium inventory are different in each controlling region, it is necessary to grasp the operational situation of the blanket for understanding the behavior of bred tritium in a blanket system. For instance, it is known from Eqs. (2)–(5) and (11) or from Figs. 6 and 7 that the residence time is proportional to the square of the grain size in the diffusion controlling region and is inversely proportional to the grain size in the surface reaction con-

trolling region. Consequently, the linear dependence of the residence time on the grain size observed in the TEQUILA and COMPRIMENT experiments indicates that their data were taken in the intermediate region nearer to the diffusion controlling region although they claimed that the surface reaction is the rate controlling step in their experiments. Then, the effect of diffusion on the residence time cannot be ignored in the operational conditions of their experiments.

The contribution of diffusion to the total inventory decreases with increase of temperature and with decrease of grain size as shown in Figs. 6 and 7. The diffusion inventory estimated using the diffusivity from Kwast et al. [10] can be ignored for the grain size smaller than several microns when the blanket temperature is higher than 573 K if He is used as the purge gas although it can be ignored even at such a low temperature as 473 K if the diffusivity by Kudo and Okuno is used. When 100 Pa H_2 is added to the He purge gas, the diffusion controlling region expands to the smaller grain size range because of decrease of the surface inventory due to the isotope exchange reaction between H_2 and tritium on the grain surface. Accordingly, the effect of diffusion in the grain cannot be ignored even at 873 K when He with 100 Pa H_2 is applied to the blanket packed with $LiAlO_2$ pellets of 4 microns in grain size because Fig. 7 reveals that such a condition is in the intermediate region if the diffusivity from Kwast et al. [10] is applied.
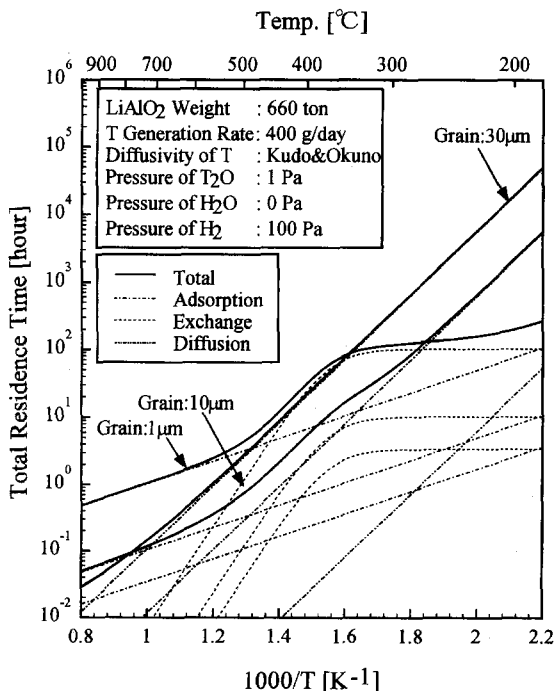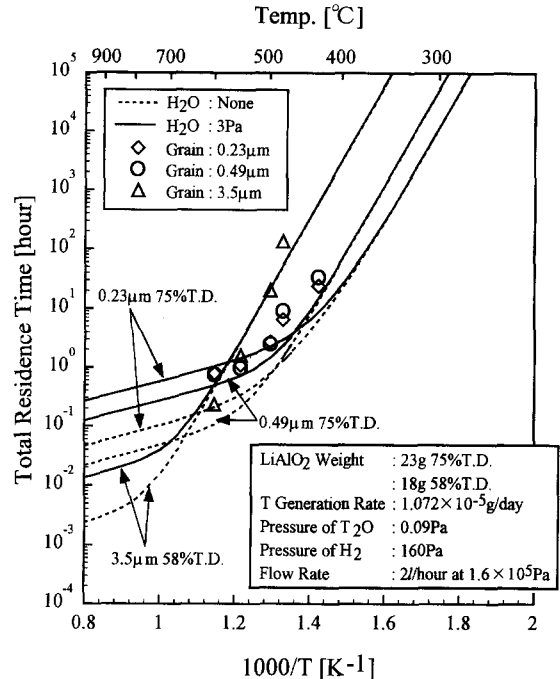

Fig. 8. Comparison of estimated values with observed residence time at in situ experiment (LILA-3).

3.3. Comparison with various in situ experiments

In the case of the in situ experiment, it is probable that some amount of water vapor got mixed with the purge gas because of insufficient dehumidification of the source gas, desorption of water from the piping surface or water formation reaction on the surface of blanket materials. Then, two isotope exchange reactions compete with each other in extraction of tritium from the grain surface. For the value of α_{ex}, however, 1.0 is used in this paper assuming that contribution from two isotope exchange reactions are same because no isotope exchange reaction rate have been reported yet.

Comparison of the estimated residence time using the model of this study with the observed total residence time by Briec et al. [11] (LILA-3) in Fig. 8 shows that a good agreement is obtained when the diffusivity obtained from the experiment by Kwast et al. [10] is used in estimation although the good agreement is not obtained when the diffusivity by Kudo and Okuno is used. It is also seen from this figure that the much better agreement is obtained when existence of some residual water vapor of several Pascals is assumed in the 'dry' purge gas. It is found by the present authors [19,20] that formation of water in the various lithium ceramics beds occurs when hydrogen isotopes are introduced to the He sweep gas. Accordingly, it is logical to consider that some water exists in the blanket purge gas even when dehumidification of the source gas is

Fig. 7. Effect of grain size on residence time for $LiAlO_2$ blanket (He + 100 Pa H_2 purge, D_T: Kwast et al. [10]).

sufficient. The tendency of their residence times are well explained by our model as shown in Fig. 8, although a simple model assuming that the diffusion in grain or a certain surface reaction is the rate controlling step cannot explain their data as a whole. The residence time by Briec et al. measured for the case when He is used as the purge gas, however, shows only a fair agreement with the estimated curve using the model of this study. It is anticipated that the system effect caused by interaction of a small amount of T_2O (HTO) released to the He purge gas with the long piping system is not fully excluded in the data processing procedure. Assumption of the residual water vapor of several pascals in He gives the better agreement also in this case.

Comparison of the estimated residence time using the model of this study with residence time reported by Alvani et al. [12] (TEQUILA-1) given in Fig. 9 also shows a good agreement when the diffusivity from Kwast et al. [10] is used and some water vapor in the purge gas is assumed.

The in situ data for $LiAlO_2$ by Werle et al. [2] (LISA-2), Briec et al. [14] (MOZART) and Alvani et al. [13] (CORELLI-2) also show fair or good agreements with estimated values in this study when the diffusivity from Kwast et al. [10] is used.

The existence of some water vapor in the 'dry' purge gas is probably unavoidable. Thus, the relative intensity between two isotope exchange reactions can give effect on the tritium inventory in the region where the surface reactions is prevailing although contributions from the

both isotope exchange reactions are supposed to be same in this study. Accordingly, quantification of the reaction rate of each isotope exchange reaction is required for development of the further discussion.

There have been reported several models to explain the tritium release curve from the blanket material under irradiation or after irradiation condition [24,25]. However, considerations about adsorption and isotope exchange reactions are missing in those models. The quantitative understanding about such properties as adsorption capacity, absorption capacity, isotope exchange capacity, adsorption rate, absorption rate, isotope exchange reaction rates and effective diffusivity of tritium in grain including the effect of irradiation defects besides reliable measurement of partial pressure of hydrogen and water vapor in the purge gas is required to evaluate the dynamical release behavior of tritium from the blanket material to the purge gas. Moreover, the system effect due to interaction of tritium with the system surface and the memory effect due to interaction of tritium with the monitor surface must be quantified to understand the release curve. It can be regrettably said that the quantitative understanding about various properties at present is not sufficient enough to give the correct prediction of tritium release curve measured by a monitor placed in the lower course of a purge gas stream.

4. Conclusions

The tritium inventory in the $LiAlO_2$ blanket under the uniform and steady state condition is estimated by considering the processes of diffusion in grain, adsorption on the grain surface and two isotope exchange reactions between H_2 or H_2O and tritium on the grain surface. The isotope exchange capacity is also considered in the estimation of this study.

Estimation using the model of this study gives a good agreement with total residence time reported in various in situ experiments for $LiAlO_2$ when the effective diffusivity evaluated in this study using the residence time reported by Kwast et al. [10] (EXOTIC-6) is applied.

It is probable that some amount of water vapor exists in the 'dry' purge gas judging from the inventory fitting in this study.
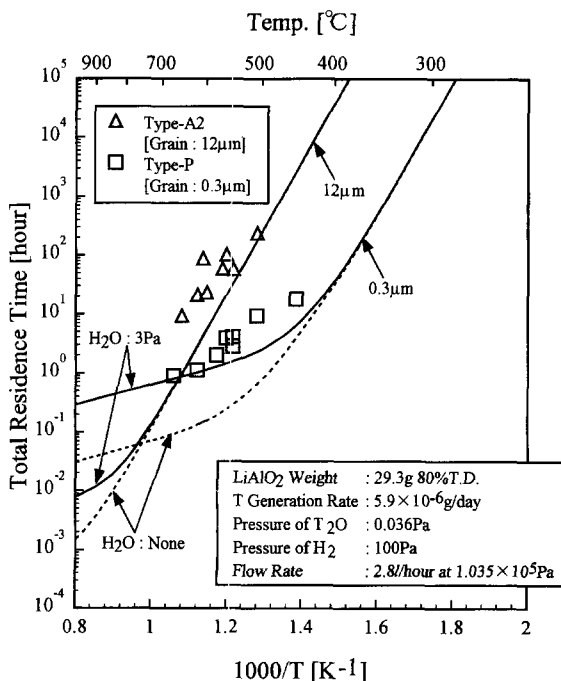

Fig. 9. Comparison of estimated values with observed residence time at in situ experiment (TEQULA-1).

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